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Spontaneous Decay and Resonance Fluorescence of an Admolecule Near
a Silver Surface with Random Roughness

by

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The spontaneous decay rate and resonance fluorescence spectrum of a molecule adsorbed near a rough silver surface are calculated. An assumed random distribution of the roughness on the surface is treated as an effective layer. The calculated decay rate is in excellent agreement with experimental measurements. The effects of surface roughness on both the spontaneous decay rate and resonance fluorescence spectrum are discussed for different cases.

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Spontaneous decay and resonance fluorescence of an admolecule near a silver surface with random roughness

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Abstract

The spontaneous decay rate and resonance fluorescence spectrum of a molecule adsorbed near a rough silver surface are calculated. An assumed random distribution of the roughness on the surface is treated as an effective layer. The calculated decay rate is in excellent agreement with experimental measurements. The effects of surface roughness on both the spontaneous decay rate and resonance fluorescence spectrum are discussed for different cases.

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I. Introduction

Fluorescence and light scattering from atoms or molecules near solid surfaces have aroused much interest since the perfection of the fatty-acid monolayer assembly technique[1] has led to a series of successful measurements of the lifetime of excited molecules near metal surfaces.[2] Despite other possible effects,[3] the role of surface roughness has been recognized to be prominent in various surface processes[4] ever since the discovery of the surface-enhanced Raman effect[5] in 1974. Processes such as photolysis and photochemical degradation have been studied in great detail by theoretical models including both localized[6] and extended[7] surface structures. It has become clear that the ultimate outcome for the enhancement of photoabsorption and resonance fluorescence processes generally depends on two competing factors. One is the enhanced surface electromagnetic field, and the other is the surface-induced decay rate of adatoms.[4,8]

Effects of the surface roughness on the spontaneous decay rate of adsorbed molecules has been investigated both theoretically[9] and experimentally.[10] It is found that the decay rate increases due to the presence of surface roughness. As a matter of fact, the decay rate of pyrazine molecules near a silver surface is measured to be about five times larger than the theoretically expected value which is calculated for a flat smooth surface. Such a tremendous increase, as has been suggested recently,[11] can be attributed to the roughness of the silver surface by assuming periodically roughened metallic surfaces.

Resonance fluorescence of adatoms at rough metallic surfaces, on the other hand, has also been studied[12] by considering the roughness as hemispherical protrusions on perfectly conducting solid surfaces. We consider, in this article, the spontaneous decay rate and resonance

fluorescence spectrum of a molecule adsorbed near a rough silver surface. The roughness is assumed to be small clusters of atoms, that may or may not be silver, distributed randomly on the surface, and is treated as an effective layer[13] with its effective optical constants modeled by the Maxwell-Garnett theory.[14] This treatment has been proven very successful in the investigation of optical excitation of surface plasma waves along rough silver surfaces. Our results are found to be in excellent agreement with experiments.

II. Theory

Consider a molecule with two levels $|+>$ and $|->$ separated by a distance $\hbar\omega$. It is adsorbed near a bulk silver with rough surface. The molecule is driven by a monochromatic laser field with

$$E(t) = \frac{1}{2}(Ee^{i\omega_o t} + E^\star e^{-i\omega_o t}) \ . \tag{1}$$

We can apply the surface-dressed optical Bloch equation[15,16] (SBE) to describe the interaction process. The SBE are given by[16]

$$\frac{d}{dt}\begin{bmatrix}<S^+>\\<S^z>\\<S^->\end{bmatrix} = \begin{bmatrix}i(\Delta+\Omega^S) - \gamma & i\Omega & 0\\ i\Omega^\star/2 & -2\gamma & -i\Omega/2\\ 0 & -i\Omega^\star & -i(\Delta+\Omega^S) - \gamma\end{bmatrix}\begin{bmatrix}<S^+>\\<S^z>\\<S^->\end{bmatrix} - \begin{bmatrix}0\\\gamma\\0\end{bmatrix} \ . \tag{2}$$

The notation is as follows. The adatom with a transition frequency ω is located at a distance d away from the surface of silver. The matrix element of the electric dipole moment operator is denoted by $|p|$, and E and ω_c are the amplitude and frequency of the external laser field, respectively. The

detuning is $\Delta = \omega - \omega_0$, and the Rabi frequency is $\Omega = |p|E$. The transition probability amplitude is proportional to the projection operators defined by

$$S^+ = |+\rangle\langle-| \quad .$$

$$S^z = \frac{1}{2}(|+\rangle\langle+| - |-\rangle\langle-|) \tag{3}$$

$$S^- = |-\rangle\langle+| \quad .$$

The total decay rate of the adatom can be written as

$$\gamma = \gamma^0 + \gamma^s \quad , \tag{4}$$

where γ^0 is the decay rate in the absence of the substrate,

$$\gamma^0 = \frac{2}{3}\sqrt{\epsilon}\ |p|^2\omega^3/c^3 \quad , \tag{4a}$$

and

$$\gamma^s = |p|^2\ \mathrm{Im}\ f(d) \tag{4b}$$

is the decay rate induced by the surface. The frequency shift of the spontaneous radiation due to the surface is

$$\Omega^s = |p|^2\ \mathrm{Re}\ f(d) \quad , \tag{5}$$

and the function $f(d)$ is introduced only for convenience[12] and is determined by[16]

$$E_R = |p|f(d)S^- = pf(d) \quad . \tag{6}$$

where E_R is the component of the reflected field $\vec{E}_R$ in the direction of $\vec{p}$.

Equation (2) agrees with the result from linear response theory[17] when the adatom is taken as a harmonic oscillator. When the intensity of the incident laser is weak, it is more probable to find the molecule in its lower state. Thus we can take $\langle S^z \rangle \approx -\frac{1}{2}$. The SBE (2) can then be linearized and solved. With the initial condition[16] $\langle S^z(0) \rangle = -\frac{1}{2}$, we find the solution

$$\langle S^z(t) \rangle = |\Omega|^2 [1 + e^{-2\gamma t} - 2e^{-\gamma t}\cos(\Delta + \Omega^s)t]/4|z|^2 - \frac{1}{2} \quad , \tag{7}$$

where $z = \gamma + i(\Delta + \Omega^s)$. By making use of the regression theorem for correlation functions,[18] we find from the SBE the well-known results of the incoherent resonance fluorescence spectrum,[18,19]

$$\tilde{g}(\nu) = \frac{1}{2}|\Omega|^4 \gamma(D^2 + \frac{1}{2}|\Omega|^2 + 4\gamma^2)/(\frac{1}{2}|\Omega|^2 + |z|^2)(x^2 + y^2) \quad , \tag{8}$$

where $D = \nu - \omega_o$, $x = 2\gamma(\frac{1}{2}|\Omega|^2 + |z|^2 - 2D^2)$, and $y = D(|\Omega|^2 + |z|^2 + 4\gamma^2 - D^2)$.

It is seen from Eqs. (4)-(8) that both the spontaneous emission properties and the resonance fluorescence spectrum depend sensitively on the surface-reflected field $\vec{E}_R$ which, in turn, depends on the optical constant of silver and the structure of the silver surface. In these equations, we have regarded the molecule as an emitting dipole. The emitted field is reflected back from the surface and is coupled to the radiation dipole whose dynamical behavior is therefore totally changed. To calculate the reflected field, we must take into account the roughness of the silver surface. This is treated

in a similar fashion as is discussed in Ref. 13. The roughness is assumed to be on a very fine scale which is much smaller than the wavelength of the relevant light. The scattering of light due to the roughness is then very small and consequently the roughness can be modeled by introducing an effective layer on the silver surface. The optical constant ϵ_2 of this layer is evaluated by the Maxwell-Garnett theory

$$\epsilon_e = \epsilon[\epsilon_s(1+2q) + 2\epsilon(1-q)]/[\epsilon_s(1-q) + \epsilon(2+q)] , \tag{9}$$

where ϵ is the dielectric constant of the medium containing the molecule, ϵ_s stands for the complex dielectric constant of the silver substrate, and q represents the volume fraction of silver in the effective thin layer. It is noted that the maximum roughness corresponds to q = 0.5.

The reflected electric field at the dipole can be found by classical electromagnetic field theory.[16,20] The results are, with $x_o = \omega/c$,

$$E_R^{\parallel} = \sqrt{\epsilon}\kappa_o^3 \, p \int_0^{\infty} d\kappa \; \kappa\mu^{-1}(R_{\perp} + \mu^2 R_{\parallel}) \tag{10}$$

when the dipole moment $\vec{p}$ is parallel to the surface and

$$E_R^{\perp} = -i\sqrt{\epsilon} \; \kappa_o^3 p \int_0^{\infty} d\kappa \; \kappa^3\mu^{-1}R_{\parallel} \tag{11}$$

when $\vec{p}$ is normal to the surface. Here we have introduced $\mu = \sqrt{1 - \kappa^2}$ and

$$R_{\perp,\parallel} = \frac{R_{\perp,\parallel}^m + R_{\perp,\parallel}^e}{1 + R_{\perp,\parallel}^m R_{\perp,\parallel}^e} e^{i2\mu\hat{d}}$$

with

$$R_{\parallel}^{m} = \frac{\epsilon \mu_e - \epsilon_e \mu}{\epsilon \mu_e + \epsilon_e \mu}$$

(13a)

$$R_{\perp}^{m} = \frac{\mu - \mu_e}{\mu + \mu_e}$$

(13b)

$$R_{\parallel}^{e} = \frac{\epsilon_e \mu_s - \epsilon_s \mu_e}{\epsilon_e \mu_s + \epsilon_s \mu_e} \, e^{i2\mu_e \hat{d}_e}$$

(14a)

$$R_{\perp}^{e} = \frac{\mu_e - \mu_s}{\mu_e + \mu_s} \, e^{i2\mu_e \hat{d}_e}$$

(14b)

$$\mu_e = \sqrt{\epsilon_e/\epsilon - \kappa^2}$$

(15a)

$$\mu_s = \sqrt{\epsilon_s/\epsilon - \kappa^2}$$

(15b)

$$\hat{d} = 2\pi\sqrt{\epsilon}d/\lambda$$

(16a)

and the thickness of the effective layer

$$\hat{d}_e = 2\pi\sqrt{\epsilon} \, d_e/\lambda \quad .$$

(16b)

Substituting (10) and (11) in (6), we can rewrite γ and Ω^s in (4) and (5) for dipole orientations parallel and normal to the surface,

$$\gamma_{\parallel} = 1 + \frac{3}{4} \, \text{Re} \int_0^{\infty} d\kappa \, \kappa \, (R_{\perp} + \mu^2 R_{\parallel})/\mu$$

(17a)

$$\gamma_\perp = 1 - \frac{3}{2} \, \text{Re} \int_0^\infty \kappa^3 d\kappa \ R_\parallel / \mu \qquad (17b)$$

$$\Omega_\parallel^s = - \frac{3}{4} \, \text{Im} \int_0^\infty \kappa d\kappa \ (R_\perp + \mu^2 R_\parallel) / \mu \qquad (18a)$$

$$\Omega_\perp^s = \frac{3}{4} \, \text{Im} \int_0^\infty \kappa^3 d\kappa R_\parallel / \mu \qquad (18b)$$

where we have taken the unit $\gamma^0 = 1$.

III. Results and discussion

The integrals involved in (17) and (18) can only be evaluated numerically. We choose $\epsilon = 1.7$, $q = 0.2$ and $d_e = 10$ Å in our calculation. In the neighborhood of the plasmon resonance, we consider two wavelengths, $\lambda = 3800$ Å and 3200 Å, for transitions of the adsorbed molecule. The dielectric constants of silver corresponding to these wavelengths are[10,21] $\epsilon_s(\lambda = 3800) = -3.16 + 0.29i$ and $\epsilon_s(\lambda = 3200) = 0.50 + 0.04i$, respectively. The roughness effect on the wavelength dependence of the decay rate is also investigated for the whole wavelength range.

We first consider the decay rate of the adsorbed molecule as a function of its distance d from the surface. In Figs. 1 and 2, the results computed from (17) are plotted for the two wavelengths. It is clearly observed that for $\lambda = 3800$ Å, the surface roughness enhances the decay rate whether the dipole orientation is parallel or normal to the surface. On the contrary, the roughness prolongs the lifetime of the excited molecule for $\lambda = 3200$ Å. Similar conclusion has been reported in Ref. 11 in which the more special case of periodically roughened metallic surface is considered. Our results are

shown in Fig. 3 in excellent agreement with experimental data.[10] The dashed line representing results calculated for a smooth surface is also plotted for comparison purposes. Therefore, surface roughness plays a very important role indeed and must be included in the treatment of any optical problem involving absorption.

To find how the effect of the roughness on the decay rate varies with the wavelength, we have computed the ratio R of the decay rate for a rough silver surface to that for a flat one as a function of the wavelength when the molecule is located very close to the surface. It is found that in the neighborhood of the surface plasmon resonance, the energy transfer from the adatom to the silver substrate is greatly enhanced due to the roughness. The results are depicted in Fig. 4. It is clearly seen that the roughness has very little effect on the decay rate for long wavelengths, say, $\lambda \geq 6000$ Å. In the shorter wavelength region, say, $\lambda \leq 3300$ Å, the effective layer of roughness that absorbs less energy plays a relatively important role in reducing the energy transfer. Such phenomena can be understood in the following manner. In the region where the radiation wavelengths are far away from the plasmon resonance, the energy transfer process depends mainly upon the imaginary part of the dielectric constant of the substrate. In other words, it depends on the energy absorption rate. For silver, $Im\epsilon_s$ becomes smaller as the wavelength becomes longer. Since the effective layer of surface roughness has a screening effect on the energy transfer, stronger screening is expected when $Im\epsilon_s$ is larger or when the wavelength is shorter.

We have also investigated the variation of the roughness effect with the distance by calculating the ratio R as a function of the distance d. As expected, this effect increases as d decreases. Thus, whether the surface roughness enhances or suppresses the spontaneous emission rate depends on both

λ and d. Figure 5 shows a typical case in which we have chosen a wavelength around the surface plasmon resonance in the short wavelength region. It is observed that the "screening effect" of the roughness layer dominates (R < 1) for small d, and R > 1 for large d. Although the results shown in Figs. 4 and 5 are calculated for the normal orientation of the adatomic dipole moment, we have obtained similar conclusions for the parallel orientation.

We now turn our attention to the interaction of an external driving field with the molecule. In what follows, we only consider the parallel orientation of the dipole moment for simplicity. The normal orientation can be treated in the same fashion, and no qualitatively different conclusion is expected. As we have noted above, we use γ^o as the unit for all quantities with the dimension t^{-1}. The behavior of the mean molecular inversion as a function of time is calculated from (7) and is shown in Fig. 6. Strong dependence of the results upon the wavelength is obvious as the only difference in (a) and (b) is in the wavelength. This qualitatively different appearance of the curves is primarily due to the energy transfer from the excited molecule to the silver surface. In the case (a), such transfer is weak and the Rabi oscillation of $S^z(t)$ is evident. The surface roughness has less effect in this case. The situation is totally different in case (b). The energy transfer is so strong that Rabi oscillation can hardly occur and the system goes into steady-state right after the interaction starts. It is also observed that the influence of the surface roughness is very important in this case.

As to the incoherent part of the resonance fluorescence spectrum, we report in Fig. 7 some of our results computed from (8) for (a) $\lambda = 3200$ Å and (b) $\lambda = 3800$ Å. Once again, the influence of surface roughness depends strongly upon the wavelength. While the two sidebands of the spectrum appear

to be completely suppressed in (b) by the surface roughness, the spectrum does not show much quantitative changes due to the roughness in (a).

Acknowledgments

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Figure captions

1. $\log\gamma$ as a function of $\log d$ for $\lambda = 3800$ Å for the cases of a rough surface (solid line) and smooth surface (dashed line). The dipole moment is oriented (a) normal to the surface and (b) parallel to the surface.

2. Same as in Fig. 1 except that $\lambda = 3200$ Å.

3. A comparison of theoretical and experimental results for the case of parallel dipole orientation. The dashed line representing smooth-surface results is plotted only to show the importance of the roughness effect.

4. The ratio R of decay rates as a function of the wavelength λ for the silver substrate. The distance is d = 20 Å. The horizontal line represents R = 1.

5. R as a function of d for silver substrate with $\lambda = 3317$ Å. The horizontal line represents R = 1.

6. Time evolution of the quantity $(<S^z> + \frac{1}{2})/|\Omega|^2$ for d = 125 Å and $\Delta = 0$. The dipole moment is parallel to the surface. The solid (dashed) line represents results calculated for the rough (smooth) surface. (a) $\lambda = 3200$ Å, (b) $\lambda = 3800$ Å.

7. Resonance fluorescence spectrum for the dipole moment parallel to the surface and $|\Omega| = 50$, $\Delta = 0$, d = 125 Å. The solid (dashed) line represents rough (smooth) surface results. (a) $\lambda = 3200$ Å, (b) $\lambda = 3800$ Å.
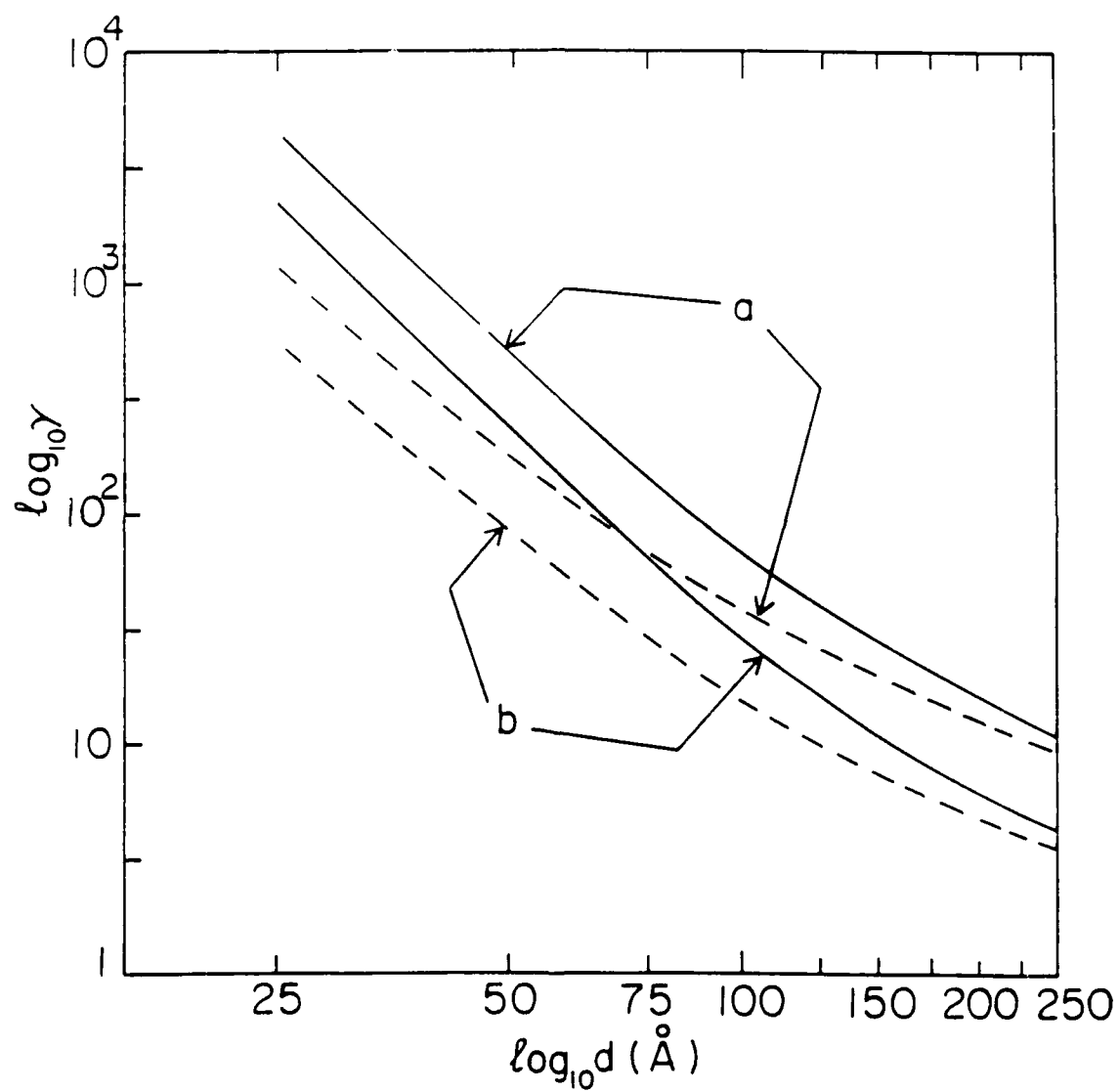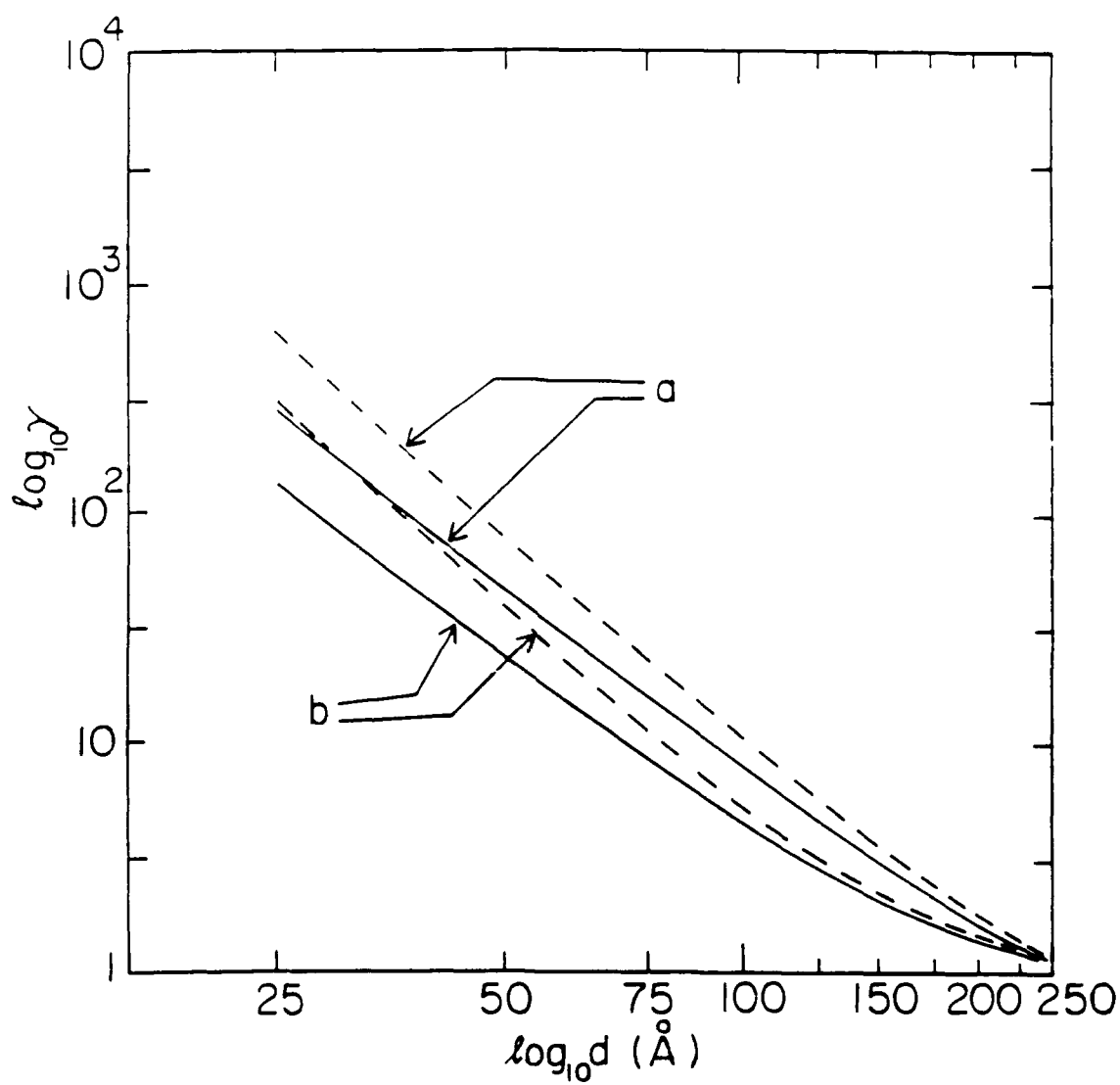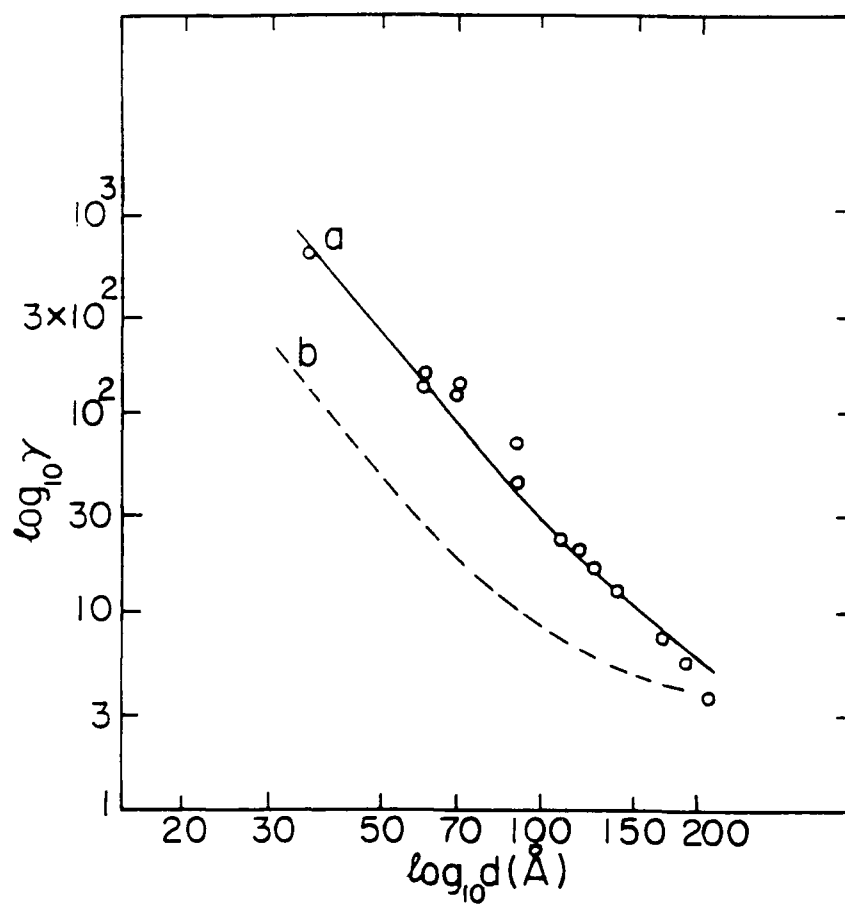
Fig 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

(a)

(b)

$(\langle S^z \rangle + \frac{1}{2})/|\Omega|^2 \ (10^{-3})$

2

1

0

0.5

1.0

t

Fig. 7

(a)

0.1

-60   0   60

$\widetilde{g}(\nu)$

0.1   (b)

-60   0   60

D
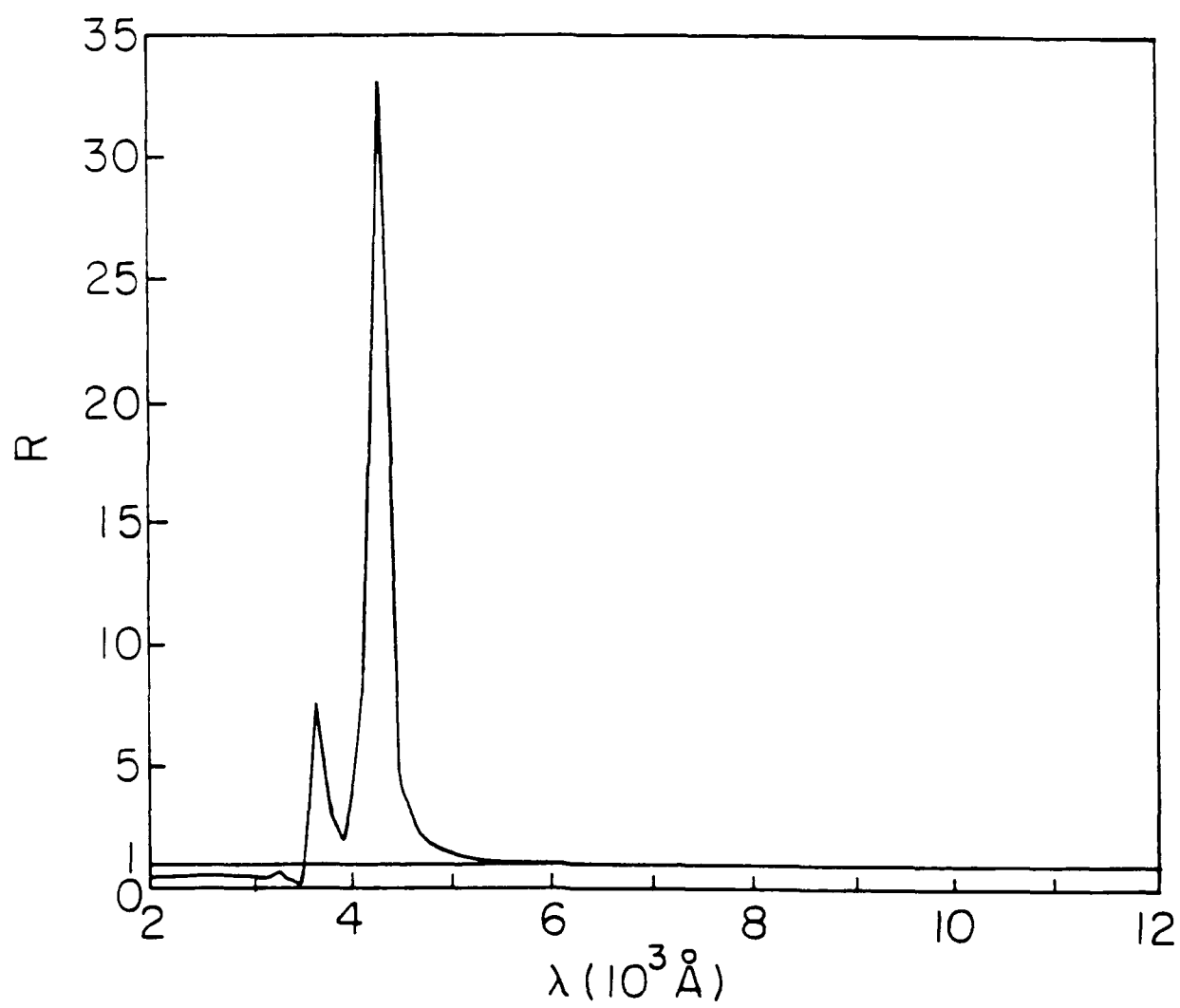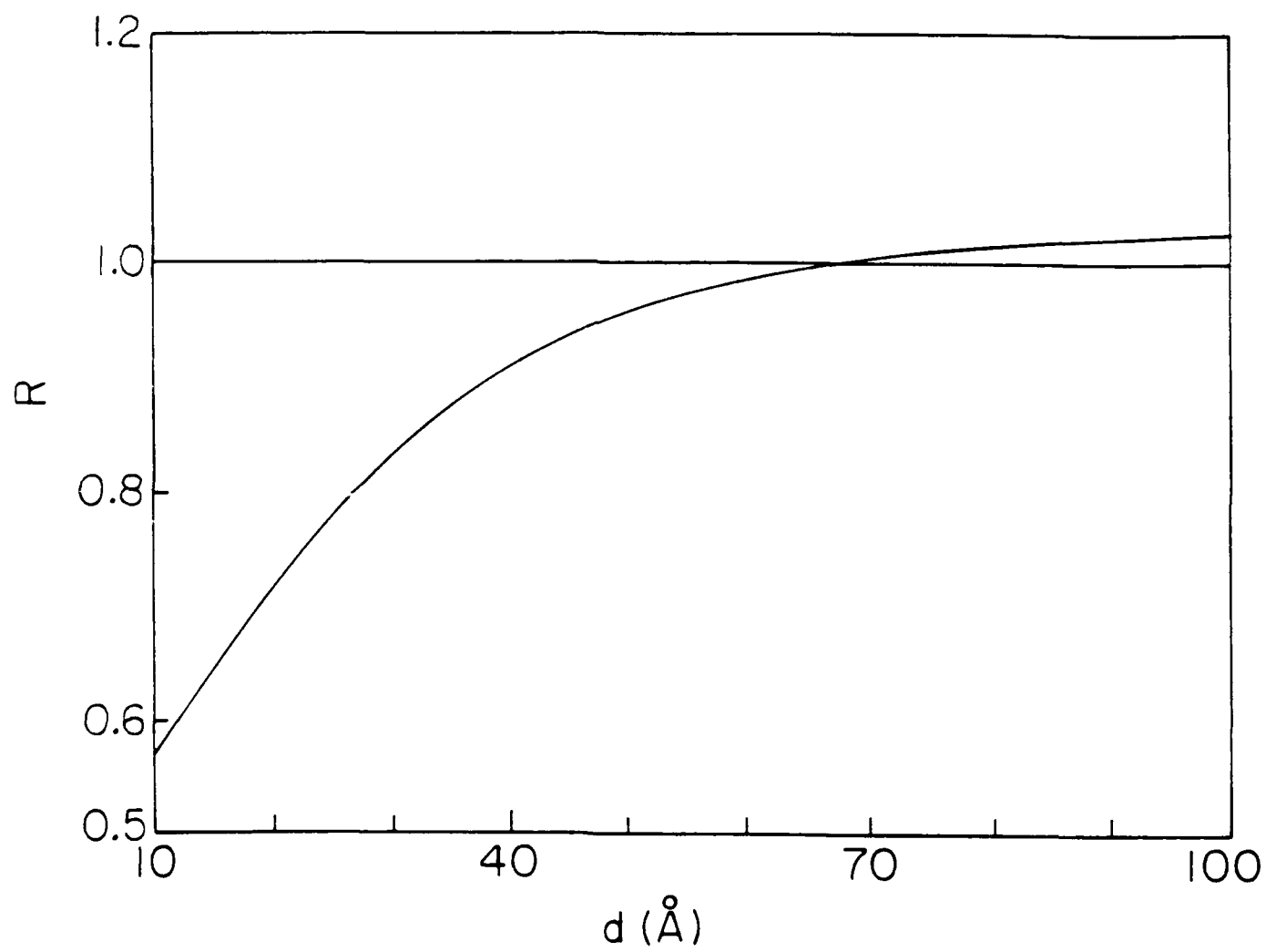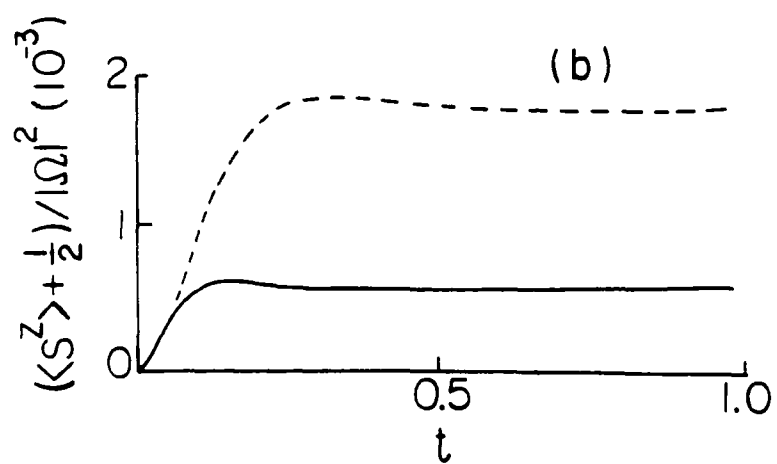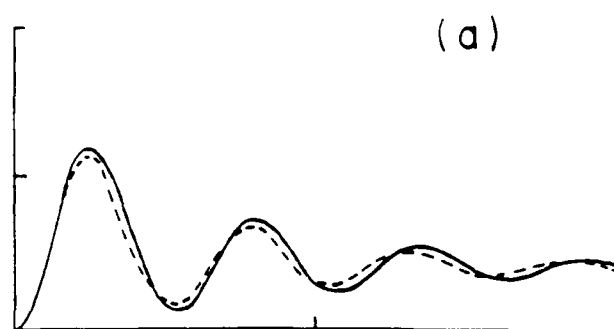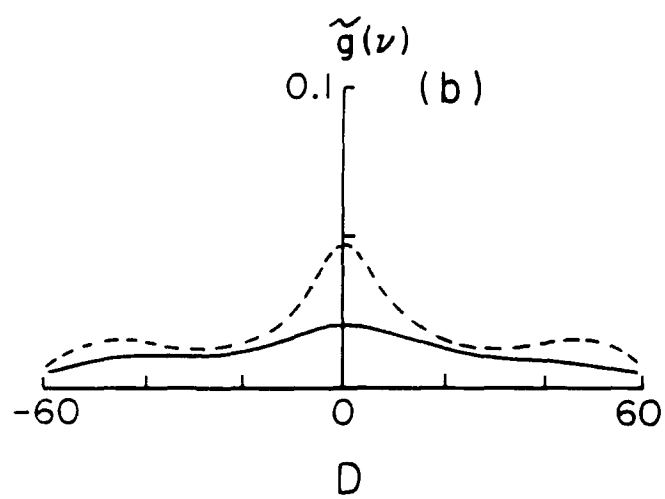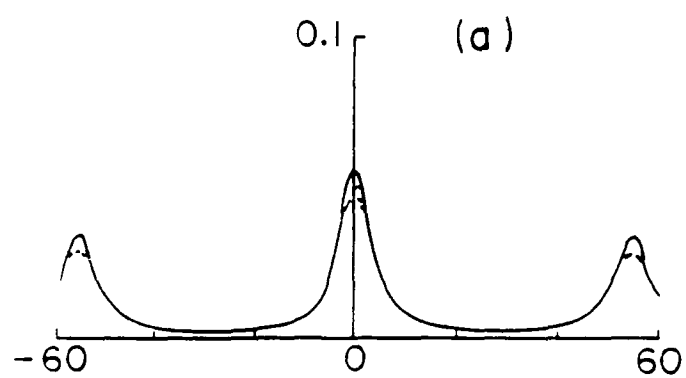
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7

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